OBSERVATION OF LATTICE INSTABILITY IN $K_{0.3}MoO_3$ BY ION-CHANNELING TECHNIQUES

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We have measured the backscattering yields of 1.00MeV He^+ ions for quasi one dimensional conductor $K_{0.3}MoO_3$ in the region from room temperature to 100K. The temperature dependence of the backscattering yields exibits the softening of a certain phonon mode. The enhancement of the backscattering yields was observed when the applied electric field for the specimens was exceeded the threshold value for the onset of nonlinear conduction in this material. The possible origin of the enhancement is discussed.

INTRODUCTION

A remarkable property of channeled ions through single crystal is their high sensitivity to distingush small lattice displacement from regular lattice sites up to the order of 0.1Å. From the minimum yield of ion-backscattering, χ_{min}, the rms displacement of atom due to thermal vibrations can be derived. These properties of channeled ions suggest potential application to investigate of crystalline phase transition [1]. However, there has been no report concerning with the application of channeled ions to study the phase transition in quasi one dimentional conductors.

In this paper, we report the first attempt of the application of the channeling technique for quasi one dimensional conductor $K_{0.3}MoO_3$ [2]. A large single crystal of $K_{0.3}MoO_3$ is now available and this enable us to carry out the experimental investigation of the response of channeled ions to the Peierls distortion and the sliding of charge density waves.

EXPERIMENTAL

Blue bronze $K_{0.3}MoO_3$ single crystals used in this experiment were grown at the Institute of Solid State Physics, University of Tokyo. We have prepared several specimens from the large crystal by cleaving along ($\bar{2}01$) plane.

A standard arrangement for ion-backscattering with an annular solid -state detector was used in this experiment. The specimen was mounted on a three-axis goniometer in the scattering chamber and the positions were controlled digitally. He^+ ions were accelerated to 1.00MeV by a Van de Graaff accelerator and the beam energy was stabilized by using feedback from slits in the beam line. The diameter and the divergence of the He^+ beam were 1.0 mm and 0.03°, respectively.

The specimen was positioned so that an axial channel which is perpendicular to the b axis was precisely aligned parallel to the beam direction at room temperature. The specimen was slowly cooled down to around 100K (below the metal-nonmetal transition temperature) and temperature dependence of the backscattering yield was measured. The cooling rate was maintained with 1K/min and temperature was stabilized within ±0.05K.

At 104K, we have measured the dc current-voltage characteristics of the specimen using 4-probe method. And the effect of the charge density wave motion on χ_{min} was measured as a function of current through the specimen.

RESULT AND DISCUSSION

1. Temperature dependence of χ_{min}

A typical backscattering energy spectra of $K_{0.3}MoO_3$ in room temperature are shown in Fig. 1. In order to minimize the radation damage of the specimen, we have adopted the larg energy window designated in the figure. From this figure, it is seen that the yield of He^+ backscatterd ions are mainly due to the collisional interaction with Mo atoms.

The temperature dependence of χ_{min} is shown in Fig. 2. Here, χ_{min} is normalized by the yield among the random directions. χ_{min} exhibits slow increase as temperature was decreased from room temperature. As temperature approaches 180K, which is the normal-incommensurate charge density wave(CDW) transition temperature, T_{ic} , in this material, the increase in χ_{min} becomes more rapidly and finally it converges to the finite value in the region where

$T \leq T_{ic}$.

According to Barrett [3], χ_{min} for monoatomic lattice is expressed as a function of the vibrational amplitude of the constituent atom as followes;

$$\chi_{min} = 3.0n\pi(\Sigma|U_q|^2> \tag{1}$$

where n is the areal density of the atom and $<|U_q|^2>^{1/2}$ is the rms displacement of the atom in mode q, in plane normal to the channeling axis. Equation(1) indicates that χ_{min} is proportional to the Debye-Waller factor of the specimen.

Let us consider the effect of some particular soft phonon modes on χ_{min} under the simplified assumptions. It is assumed that a particular phonon modes start to soften at $T=T_0$ and become unstable at $T=T_{ic}$; their frequencies change from $\omega^2(\vec{q}) = \omega_0^2(\vec{q}')$ to $\omega^2(\vec{q}) = D_1(T - T_{ic}) + D_2|\vec{q} - \vec{q}'|^2$ for $|\vec{q} - \vec{q}'| < q_c$, where $\vec{q}'$ is the wave vector of the soft mode, $\omega_0^2(\vec{q}')$ and q_c are temperature-independent constants, $D_1 = \omega_0^2(\vec{q}')/(T_0-T_{ic})$, and $D_2 = \omega_0^2(\vec{q}')(T_0-T)/(T_0-T_{ic})q_c^2$.

χ_{min} in this case is given by the sum of the term χ^*_{min} due to the soft phonons and term A^* due to the other phonons and lattice defects which are assumed to be independent of temperature. The above assumption seems resonable, because χ_{min} in various single crystals which exhibit no phase transition shows very small variation in the present temperature region;

$\chi_{min} = \chi^*_{min} + A^*$, and χ^*_{min} is expressed as [4]

$$\frac{\chi^*_{min}}{T} = \text{const.}\left\{\frac{1}{1-x} - \frac{1}{3}\left(\frac{x}{(1-x)^3}\right)^{1/2}\tan^{-1}\left(\frac{1-x}{x}\right)^{1/2}\right\} \tag{2}$$

Here, we put $x=(T-T_{ic})/(T_0-T_{ic})$.

The expression of actual χ_{min} of $K_{0.3}MoO_3$ is more complex due to the finite contribution of O and K atoms to Mo atomic potential. However, as mentioned earlier, the main contribution to the backscattering yields comes from the interaction of He^+ ions with Mo atoms, Eqs.(2) can still be used as guiding principle for serach the response of χ_{min} to the exsistence of soft phonon modes.

The dotted line in Fig. 2 indicates the theoretical curve where we assume T_0 is equal to 300K and the constant prefactor in Eq.(2) is determined so that the experimental χ^*_{min} values between 240 and 300K show the best fit to the theory. The theoretical curve indicates the qualitative feature of χ^*_{min}. Qnantitatively, the theoretical curve shows large deviation from the experimental values near T_{ic}. In order to explain the actual χ^*_{min} in $K_{0.3}MoO_3$ quatitatively, more detailed theory which includes the ansotropy of phonon dispersion is necessary.

χ_{min} in Fig. 2 shows slow increase in the intermediate region between 225 to 200K and again increases rapidly as temperature approaches to T_{ic}. The above rapid rise in χ_{min} near T_{ic} is considered to be related the fluctuation of short range order in the soft phonons near T_{ic}.

In the region where $T < T_{ic}$, χ_{min} seems to be constant and the effect of the amplitude mode or phase mode on χ_{min} is not observed. The reason is not clear at present.

In the incommensurate states in $K_{0.3}MoO_3$, χ_{min} remains larger than the one in room temperature, and this implys that a large number of domain structures is established in these states. Also certain lattice deffects might be introduced through the phase transition. These facts have been observed in transition-metal dichalcogenides [4].

Dumas et al. [5] have reported that the threshold electric field, above which the nonlinear conduction is observed, has a sharp maximum at about 110K. However, we have observed no anomaly in χ_{min} around the above temperature, and it seems that the above sharp maximum in the threshold electric field has no correlation with lattice distortion.

2. The enhancement of χ_{min} by the CDW motion.

We are interested in the reflection of CDW motion to χ_{min}. At first, we have measured the dc current-voltage characteristics in this material at 104K. The ohmic contacts were made with indium amalgam with heat treatment at 180°C for several hours in a vaccum furnace. The threshold electric field for the onset of nonlinear conduction is about 230mv/cm, which is shown in Fig. 3. Then,we have measured the backscattering yields as a function of current throgh the specimen, which is shown in Fig. 4. It is seen from this figure that χ_{min} is constant in the ohmic region, except the relatively large rise in χ_{min} near the threshold field. Dumas et al.[5] have observed large current spikes near the threshold field. It seems that the current spikes have some correlation with the rise in χ_{min}, although we have not measured the time-dependent current spikes and noise at the same time, and it has not been confirmed the above correlation. As the applied field is increased and the current exceeds the threshold for the nonlinear conduction, χ_{min} is increased slowly. When the applied is removed, χ_{min} returns to the intial value within the statistical error.

The above experimental fact indicates the additional lattice distortion or effective increase of rms displacement of lattice vibrations are introduced with the sliding motion of the CDW. In general, quasi one dimensional conductors are influenced easily by radiation of high-energy particles. In order to monitor the radiation damage of the specimen, the ohmic resistance of the specimen was often measured during the experiment, and fortunately we have not observed any noticeable change of the ohmic resistance The most He^+ ions are in the channeling state and this prevents the rapid deterioration of the specimen.

It might be conceivable that dislocations or mobile defects in the specimen will force to rearange through the motion of CDW. However, it is unlikely that the above arrangement has such a short relaxation time that the intial state is rapidly recovered when the applied field is removed.

The other possibility is as follows; the lattice distortion is considered to oscillate around its equilibrium distortion when the sliding motion of CDW occurs. This oscillation is due to match the phase change of the CDW, and the oscillation of lattice distortion will introduce the additional backscattering yield as a whole. Actually, the incommensurate phase consists of the CDW domain or discommensuration[6]. The movement and rearrangement of of the discommensurations are also considered to be the origin of the additional increase of χ_{min}, because the finite phase jump exists in the boundary of discommensurations and the lattice ions should follow this phase jump.

It is clear that such a picture would need further theoretical and experimental considerations.

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FIGURE CAPTIONS

Fig. 1. Typical 1.00 MeV He^+ ion backscattering spectra in $K_{0.3}MoO_3$ at room temperature.

Fig.2. The temperature dependence of χ_{min} in $K_{0.3}MoO_3$. The dashed curve indicates the estimated χ^*_{min} from Eq.(2). The constant pre-factor in Eq.(2) is determined so that the experimental values between 240 and 300K show the best fit to the theory.

Fig. 3. Voltage vs current characteristic of $K_{0.3}MoO_3$ obtained at 104K.

Fig. 4. The variation of χ_{min} as a function of current through the specimen at 104K.

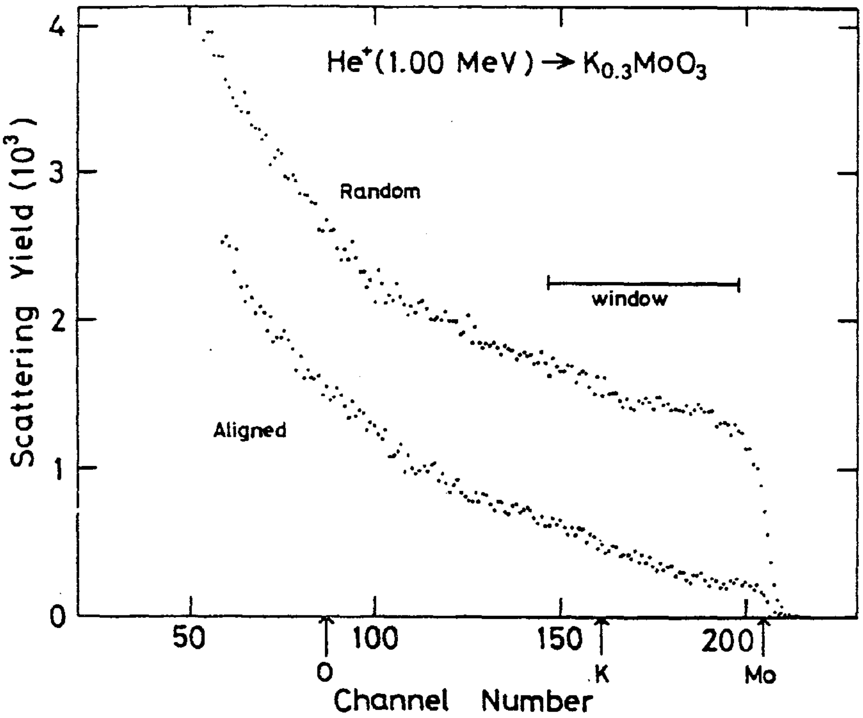
He⁺(1.00 MeV) → K0.3MoO3
Scattering Yield (10³)
Random
Aligned
window
Channel Number
O
K
Mo
4
3
2
1
0
50
100
150
200

FIG. 1.

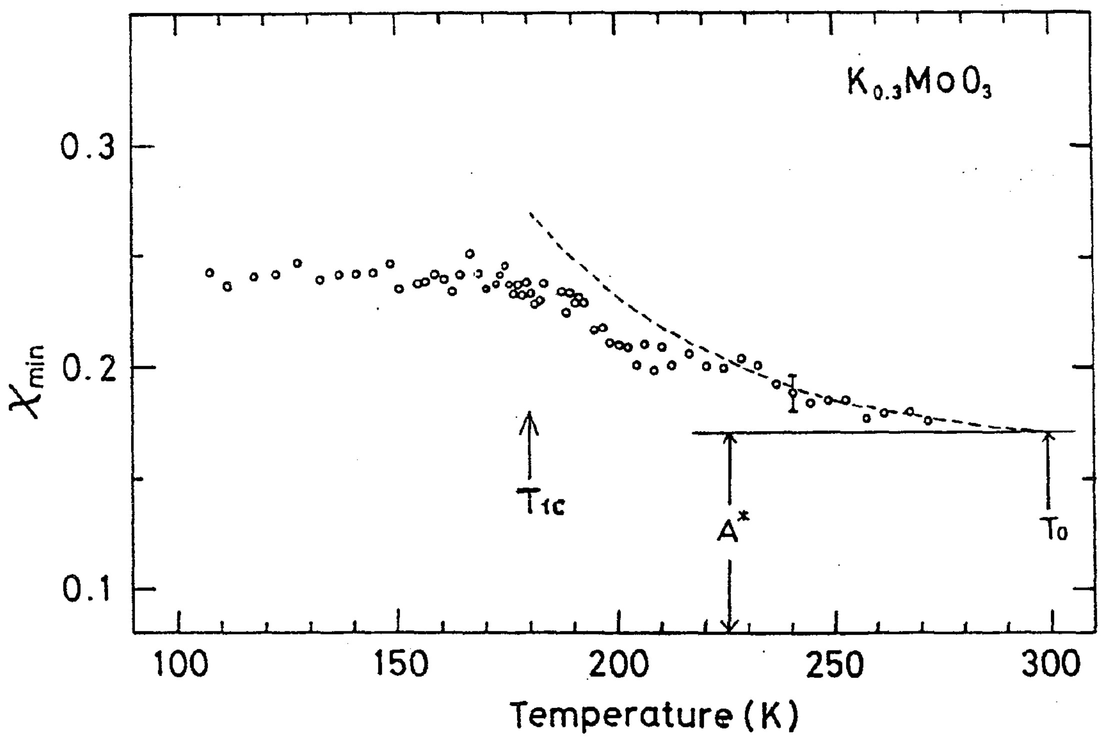
K0.3MoO3
0.3
0.2
0.1
Xmin
T1c
A*
T0
100
150
200
250
300
Temperature (K)

FIG. 2.

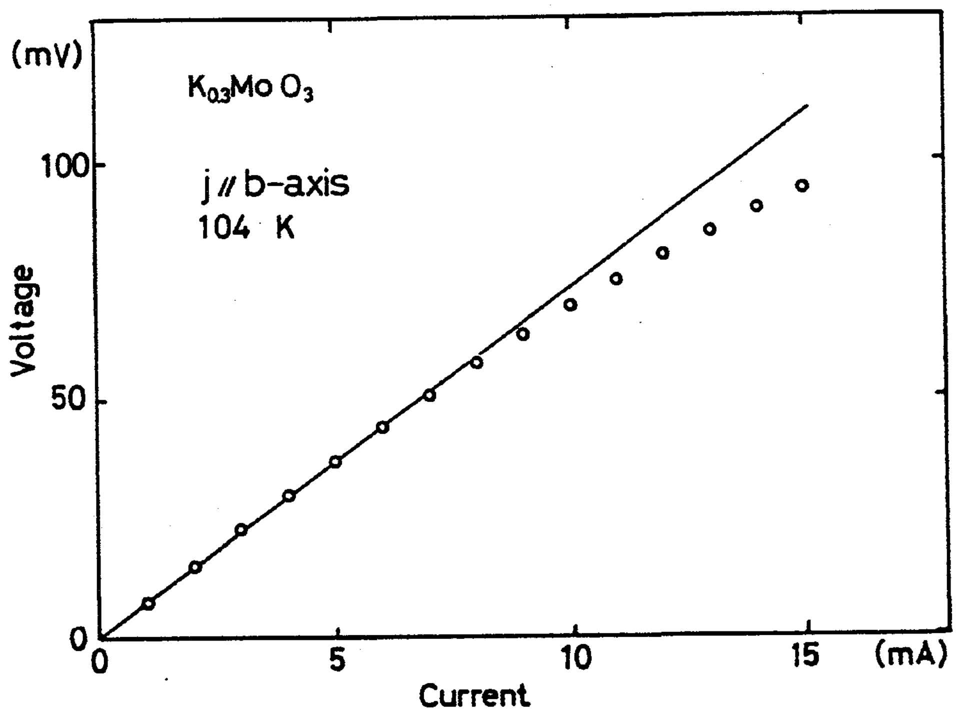
(mV)
$K_{0.3}MoO_3$
100
j // b-axis
104 K
Voltage
50
0
0
5
10
15
(mA)
Current

FIG. 3.

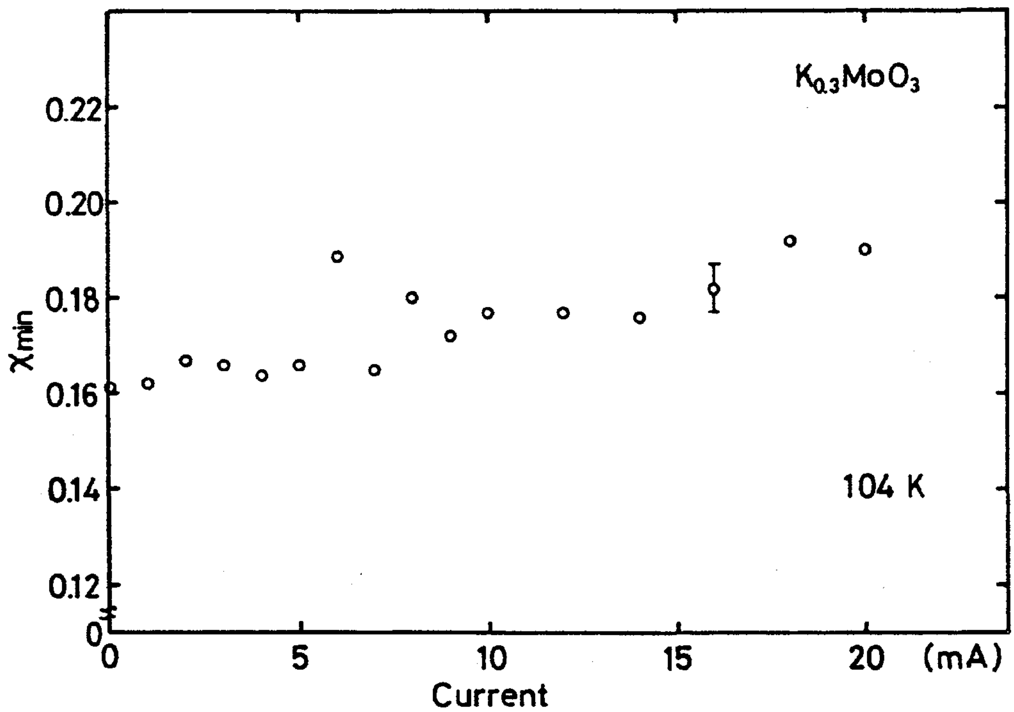
$K_{0.3}MoO_3$
0.22
0.20
0.18
χ_{min}
0.16
0.14
104 K
0.12
0
0
5
10
15
20
(mA)
Current

FIG.4.